A Flue Gas Cleanup Technology of Next Generation – Copper Oxide Process

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Abstract

Most current power plants use two separate facilities or technologies to reduce SO_x and NO_x emissions. A new flue gas cleanup technology – the Copper Oxide Process can reduce both SO_x and NO_x in a single unit. Thus this new technology has the potential to have lower costs than separate technologies when high SO_x and NO_x removals are required. The Copper Oxide Technology will also be able to turn the captured SO_x into high valued byproducts such as sulfuric acid, elementary sulfur, ammonia sulfate, and concentrated SO_2 gas. This flexibility enables local market conditions to be considered when choosing the byproduct type and can further compensate the process costs. Lastly, because the Copper Oxide Technology is a dry and regenerable process, it will not generate solid waste requiring landfills.

The Copper Oxide Technology was originally developed at the US DOE Federal Energy Technology Center (FETC). High SO_x and NO_x removals have been achieved for extended periods at FETC's Life Test Facility at 0.5 MMBTU/hr. Scale-up tests of 5 MMBTU/hr with ammonia sulfate as a byproduct are currently being conducted at the Illinois Coal Development Park by a team consisting of Sargent and Lundy, Thermo Power Corporation, and the Southern Illinois University under joint funding by the US DOE and the State of Illinois. A demonstration test at a 10 MW equivalent scale is planned for next year at a plant in Illinois.

This article will discuss the working mechanism of Copper Oxide Technology, its high sulfur and nitrogen oxide removals, technology challenges, status of development, and other issues.

Introduction

Fossil energy plays a critical role in sustaining high living standards for industrial world. Statistics shows that the mean Gross Domestic Product (GDP) per capita is approximately proportional to the mean power consumption per capita for nations at different development levels. Fossil energy contributes 80 to 85 % of the total world energy consumption, and the dependence on fossil energy is well expected to continue in the 21st century. Coal fired power plants dominate electricity generation both in the US and Korea. In 1997, coal accounted for 1856 billion kwhs or 53 percent of total generation in the US [1]. In Korea, coal plays an equally important role in electric power production. In 1997 electricity generated by fossil fuels accounted for 63 percent of total generation [2]. While coal is an essential energy source for the US and Korea, the use of coal poses significant challenges in avoiding emission of air pollutants, especially sulfur and nitrogen oxides.

The United States has achieved a significant progress in the reduction of sulfur and nitrogen oxides from coal fired power plants. The sulfur dioxide emissions from electricity generation have been reduced from 15.7 million tons in 1990 to 11.9 million tons in 1995. The sulfur emissions are expected to be reduced further according to the Clean Air Act Amendment of 1990 (CAAA 90). Beginning in 2000, the emissions from the Phase I plants (1,260 units at 110 plants) will be further tightened, and emission limits will also be set for the remaining 2,500 boilers at 1,000 plants. The US Energy Information Agency estimated that after 2000, 26.4 gigawatts of capacity – about eighty eight 300 megawatts plants will be retrofitted with scrubbers to meet the Phase II goal.

NO_x emission in the US is expected to fall significantly over the next 5 years as new legislation takes effect. It is expected that the second phase of CAAA 90 will result in NO_x reduction of 1.5 million tons between 1999 and 2000. In addition, the ozone transport rule (OTR) will take effect in 2003, which sets caps on NO_x emissions in each of 22 midwestern and eastern States during the 5-month summer season. The OTR is expected lead to total NO_x emissions reduction of 0.7 million tons between 2002 and 2003.

Currently, many power plants use two separate facilities or technologies to reduce the SO_2 and NO_x emissions. The Federal Energy Technology Center (FETC) of the US Department of Energy is sponsoring development of the Copper Oxide Technology that can remove both SO_2 and NO_x from the flue gas in a single unit. Thus this technology has the potential to have a lower cost than separate technologies when high sulfur and NO_x removals are required. The Copper Oxide Process is a dry regenerable process that has many advantages over wet scrubbers. The Copper Oxide Process will not produce landfill waste, thus avoiding concerns over the limited landfill space, increasing landfill costs, and public awareness of the environmental impact. The Copper Oxide Process also provides an effective way to use sulfur as a byproduct. The sulfur released from the process is a concentrated SO_2 stream that can be used to produce sulfuric acid, elemental sulfur, fertilizer, etc. The high value of the byproducts can partially compensate the costs for operating the process, and the flexibility in selecting byproducts could better meet local demands. The overall costs for various flue gas cleanup processes are given in Figure 1.

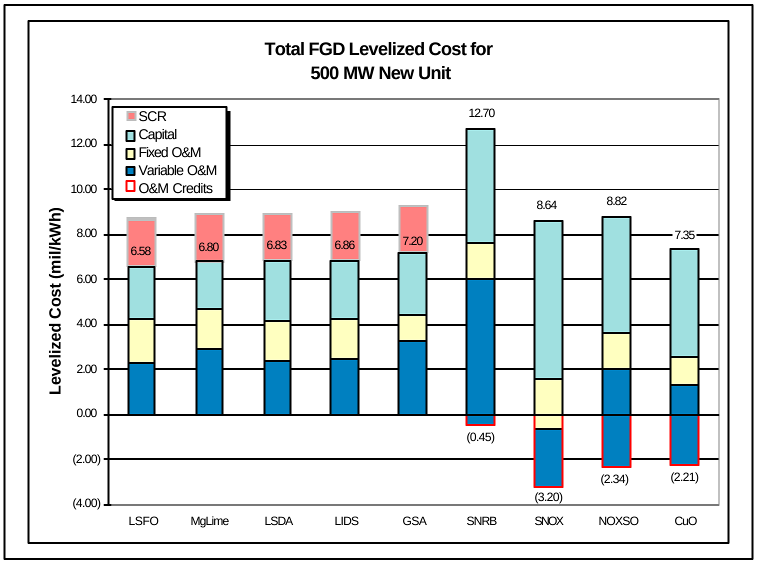


Figure 1. Total Levelized Cost for Various Flue Gas Cleanup Processes

The Copper Oxide Technology was originally developed at the Federal Energy Technology Center. High SO_x and NO_x removals have been achieved for extended periods at FETC's Life Test Facility at 0.5 MMBTU/hr. Scale-up tests of 5 MMBTU/hr with ammonia sulfate as a byproduct are currently being conducted at the Illinois Coal Development Park by a team consisting of Sargent and Lundy, Thermo Power Corporation, and the Southern Illinois University, under joint funding from the US DOE and the State of Illinois. A demonstration test at a 10 MW equivalent scale is planned to start at an ethanol plant at Pekin, Illinois next year. The performance goals, given at Table 1, are greater than 95% sulfur removal (at most 0.1-0.3 LB/MMBTU SO_2 emissions for a flue gas containing 2.8 –5.6 LB/MMBTU SO_2), greater than 90% NO_x removal, and 50% particulate removal.

Table 1. Performance Goals

Pollutant	Entering Flue Gas	Cleaned Flue Gas	Maxim Removal
SO_2	2.8 –5.6 LB/MMBTU	At Most 0.1-0.3LB/MMBTU	>95%
NO_x	0.15 – 0.6 LB/MMBTU	Less than 0.1 LB/MMBTU	>90%
Particulate	10.5 LB/MMBTU	5 LB/MMBTU[1]	50%

[1] Particulate is further removed in a bag house or precipitator to 0.01LB/MMBTU.

Working Mechanism of CuO Process

The basic reactions of the CuO process were investigated early in 1960's. McCrea *et al* [3] reported on the bench-scale evaluation of SO_2 removal using copper-impregnated alumina spheres and presented preliminary design criteria for a process to be used to treat flue gas from a 1000-MW plant burning coal with 3% sulfur.

CuO can readily react with SO_2 in the flue gas at temperatures around 650 to 750 ^oF to form $CuSO_4$. $CuSO_4$ then can be reduced to Cu by methane or other reducing gases, releasing SO_2 in a concentrated form that can be used in various processes. The regenerated sorbent is exposed to the flue gas, the elemental copper is converted to copper oxide that can be again used to react with SO_2. The main reactions can be expressed as follows.

$$CuO + SO_2 + ½\ O_2 = CuSO_4$$

$$CuSO_4 + ½\ CH_4 = Cu + SO_2 + ½\ CO_2 + H_2O$$

$$Cu + ½\ O_2 = CuO$$

An interesting feature of the process is that both $CuSO_4$ and CuO can serve as catalyst for reducing NO_x to N_2 using NH_3. By injecting NH_3 into the flue gas before it contacts CuO impregnated sorbent, both NO_x and SO_2 in the flue gas can be removed. The NO_x reduction reactions can be written as:

$$4\ NO + 4\ NH_3 + O_2 = 4\ N_2 + 6\ H_2O,$$

$$6\ NO + 4\ NH_3 = 5\ N_2 + 6\ H_2O,$$

$$2NO_2 + 4NH_3 + O_2 = 3\ N_2 + 6\ H_2O, \text{ and}$$

$$6\ NO_2 + 8\ NH_3 = 7\ N_2 + 12\ H_2O.$$

The basic schematic flow diagram is given in Fig.2. The two principal flow paths, flue gas and solid sorbent, are described. The flue gas enters the absorber where SO_2 is removed and NO_x is reduced through contact with regenerated sorbent. The clean flue gas leaves the absorber and can be discharged to a heat exchanger to recover the heat content in the flue gas. The spent sorbent then is fed to the regenerator where $CuSO_4$ is reduced to Cu using methane or other reducing gas, and SO_2 is released in the off gas stream. The regenerated sorbent is fed back to absorber and the cycle of sulfur removal is completed. NH_3 is added to the absorber to reduce NO_x to nitrogen.

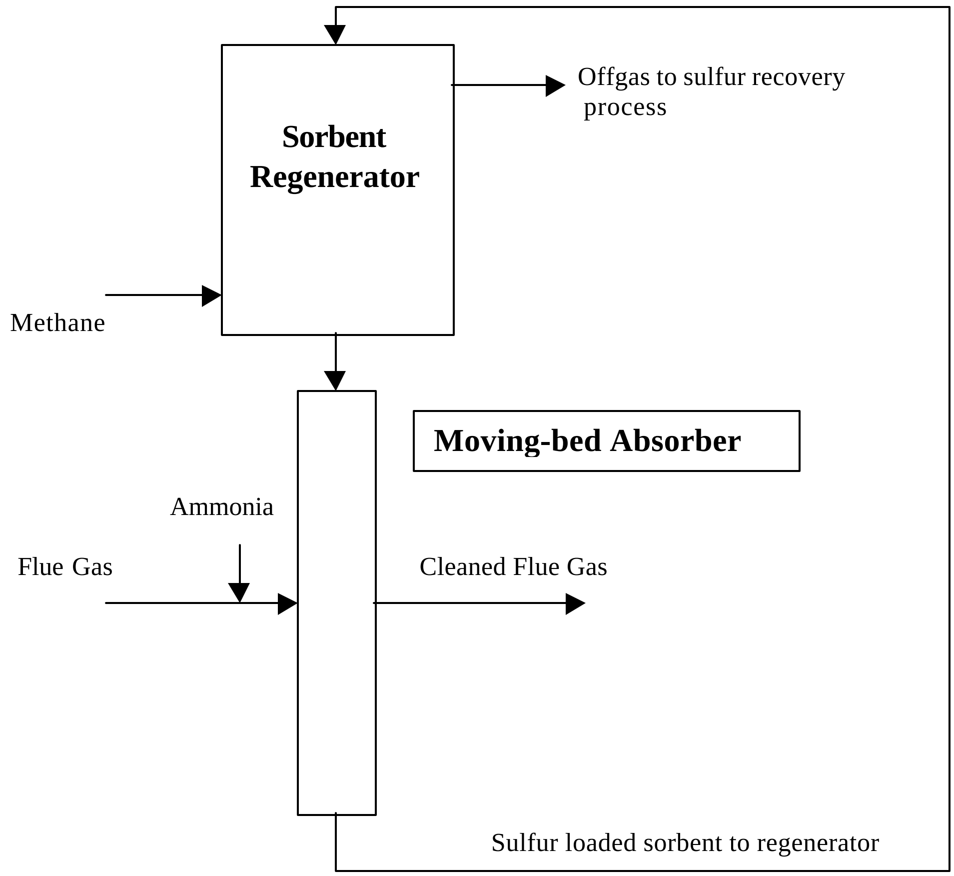


Figure 2. Schematic diagram of copper oxide process

In addition to the two main reactors, absorber and regenerator, a sorbent heater may be required after the absorber to raise the sorbent temperature for efficient regeneration.

The sorbent used here is a copper impregnated gamma alumina of 1/16” to ¼” diameter. The weight percentage of copper is around 6-7 %. If the copper content is higher than 7%, pore plugging will limit the utilization of the copper and result in degradation in the strength of the sorbent particle.

History of Development

The Copper Oxide Process has been tested in several different configurations. The original McCrea bench scale testing used a fixed bed reactor, where absorption and regeneration reactions take place alternatively in the same reactor. Shell Flue Gas Treating (SFGT) also used a fixed bed configuration. This process was tested between October 1979 to October 1980 at a pilot plant using a slipstream from a coal-fired unit at

Tampa Electric Company's Big Bend Station. Normal flue gas flow-rate to the pilot plant was 1600 Nm^3/hr (1000scfm), and flue gas was processed for a total of about 2000 hours during the program [4].

Fluidized-bed configuration testing first began at FETC in 1975 in a 6-inch diameter reactor. 75 absorption and regeneration cycles were conducted and promising results led to a scale-up tests using 3ft 4 inch x 4ft cross-section, and 12 ft high fluid absorber. The flue gas was supplied by a 500 lb/hr pulverized-coal-fired combustion test facility. Sulfur dioxide removal efficiencies in excess of 90% were readily achieved throughout the test program. Nitrogen oxide removal efficiencies over 90% were achieved at NH_3/ NO_x mole ratio of 1.0 or less [5].

FETC later moved to develop a moving bed Copper Oxide Process. The main advantage of using a moving bed versus fluid bed reactor is less sorbent attrition. Because a large amount of sorbent is required to achieve high sulfur removal efficiencies, sorbent make up costs stemming from sorbent attrition is a major cost item. Compared to the earlier fixed bed reactor configuration, the moving bed configuration can have significantly lower capital costs since multiple reactors and complex gas valves and ducting are not required. Sulfur removal efficiency and sorbent utilization is better controlled in a moving bed than a fixed bed. Furthermore, in the moving bed configuration the absorber and regenerator each can be operated at a steady optimum temperature, while this is more difficult to achieve in a fixed bed configuration.

FETC began to build a moving bed Copper Oxide Process at the Life-Cycle Test Facilities at 0.5 MMBTU/hr in 1994. The test results show that this process can meet the goals of a Department of Energy initiative to develop flue gas control technologies to remove 99% and 95% of SO_2 and NO_x, respectively [6]. In a moving bed configuration high sulfur removal can be achieved either by increasing the bed depth, or the sorbent flow rate. When the bed thickness is increased, additional layers of sorbent will remove remaining sulfur dioxide in the flue gas before it leaves the absorber. When the sorbent flow rate is increased, more clean sorbent will enter the absorber per unit of time, and this will also increase the copper to sulfur ratio and thus enhance the sulfur removal.

The extensive laboratory and pilot scale testing of the Copper Oxide Process has demonstrated that the chemistry of process is robust, and high removal of both SO_2 and NO_x is achievable. The key issues are how to scale up this process to a level of practical interest, and how to make this process economically competitive.

The first large scale demonstration of the moving bed Copper Oxide Process will be a part of the Low Emission Boiler System (LEBS) program. This DOE initiated program has the goal of developing the next generation of pulverized coal-fired boiler systems capable of meeting strict emissions limits while incorporating improved ash disposibility, reduced waste generation, reduced toxic substance emissions and increased thermal efficiency. The moving bed copper oxide process was chosen as the flue gas treatment technology for this new generation of boiler systems by a team consisting of Sargent and

Lundy, DB Riley and Thermo Power Corporation because of the following features of the system:

- High sulfur removal efficiencies,
- Simultaneous NO_x control,
- No waste production,
- Saleable sulfur byproduct, and
- High sulfur trioxide removal to facilitate low temperature heat recovery.

As part of the LEBS program, a 5 MMBTU/hr copper oxide facility was constructed at the Illinois Coal Development Park to demonstrate at a reasonable scale the component and subsystem designs which will make up a commercial scale power plant. Operation of the facility has validated key design features of the process equipment including the absorber retention screens, material handling equipment, and reactor configurations. Process chemistry has been verified using sorbent made with a lower cost alumina substrate and a lower cost production procedure. A full scale demonstration of the process equipment at a 10 MW equivalent scale is planned to start soon at an ethanol plant at Pekin, Illinois.

Modeling Efforts

In addition to a series of experimental studies, significant modeling efforts were conducted for Copper Oxide Process. Several microbalance tests were conducted at FETC for both SO_2 absorption and sorbent regeneration.

The chemical kinetic rate equation used to describe the sulfation of copper is given by Yeh *et al* [7]:

$$\frac{dx}{dt} = k(1-x)P,$$

where x =fractional conversion of copper oxide,
P= fractional partial pressure of sulfur dioxide,
t = time, h,
k = reaction rate constant, h^{-1}.

The activation energy for reaction was determined to be 20.1 kj/mol, and the frequency factor was 47,000 and 34,130 (h^{-1}) for 5% and 7% copper sorbent respectively.

The sorbent regeneration kinetics was first studied by Markussen *et al* [8]. Markussen studied reaction in a microbalance, and modeled the kinetics with a Langmuir – Hinshelwood mechanism. Using data obtained in Markussen's microbalance study, Chen and Yeh [9] further modified the kinetic model to consider the equilibrium limits exhibited in the data by introducing a reverse reaction term. The modified kinetics is written as:

$$\frac{dX_A}{dt} = \frac{k_2 P_{CH_4}}{1 + K_1 P_{CH_4} + K_2 P_{SO_2}} (1 - X_A) e^{-X_A} - \frac{k_3}{P_{CH_4}} X_A^{\;2}$$

where X_A is the sorbent conversion. P_{SO2} and P_{CH4} are partial pressure of SO_2 and CH_4. Additional improvement in Chen and Yeh's kinetic model includes the temperature dependence of rate constants of k_2, k_3, K_1 and K_2. The temperature dependence is important for modeling the regenerator at various temperatures, as the regeneration performance will deteriorate rapidly when the temperature decreases.

The reactor modeling was also actively pursued. In the case of an isothermal moving bed absorber, an analytic solution was found by Boyd and Chen for the reaction equations [10]. A numerical method that treats the whole moving bed as a collection of micro reactors was developed by Young and Yeh [12]. For isothermal reactors both the micro-reactor methods and analytic solution give a very close prediction, since both methods are based on the same equations. In a refinement to the numerical method, Thermo Power included the heat of reactions into the micro reactor elements thereby taking into account the temperature profile setup in the absorber due to the oxidation and sulfation reactions [11]. Several experimental studies show the model predictions are in good agreement with experiment data, and high sulfur removal can be achieved in moving bed absorbers.

The counter flow regenerator has also been modeled. Poor sorbent regeneration will lead to a sulfur accumulation on the sorbent, and this will cause deteriorating sulfur removal efficiency. In early tests, poor sorbent regeneration was observed and reasons were uncertain. Chen and Yeh studied modeling of the regenerator. They found that a major factor that slows regeneration reaction is the slow gas velocity in the regenerator. Unlike the fast gas flow in the absorber, the regenerating gas flows at a much slower velocity compared with the gas velocity used in the kinetics study using a microbalance. Including a gas velocity factor, the simulation model gave an excellent agreement with the FETC life cycle test results [9]. The model also shows that keeping adequate temperature is critical for having adequate reaction rate because regeneraion reaction will become very slow at relatively low temperature (see Fig.3).

Scale-up Test at the Illinois Coal Development Park

The Life-Cycle Test conducted at FETC was at 0.5 MBTU/hr, and satisfactory results have been achieved for extended period of time. A scale-up test was conducted by a team of Sargent and Lundy, Thermo Power Corporation, and Southern Illinois University at the Illinois Coal Development Park in Carbondale, Illinois. The test facility is at the 5 MMBTU/hr level, and a synthetic flue gas with a controlled SO_2 concentration was used. Originally, a sorbent heater was not included in the facility, but sulfur was accumulated on the sorbent because of inadequate sorbent regeneration. When a sorbent heater was added between the absorber and regenerator, regeneration performance was improved, and greater than 90% SO_2 removal has been achieved.

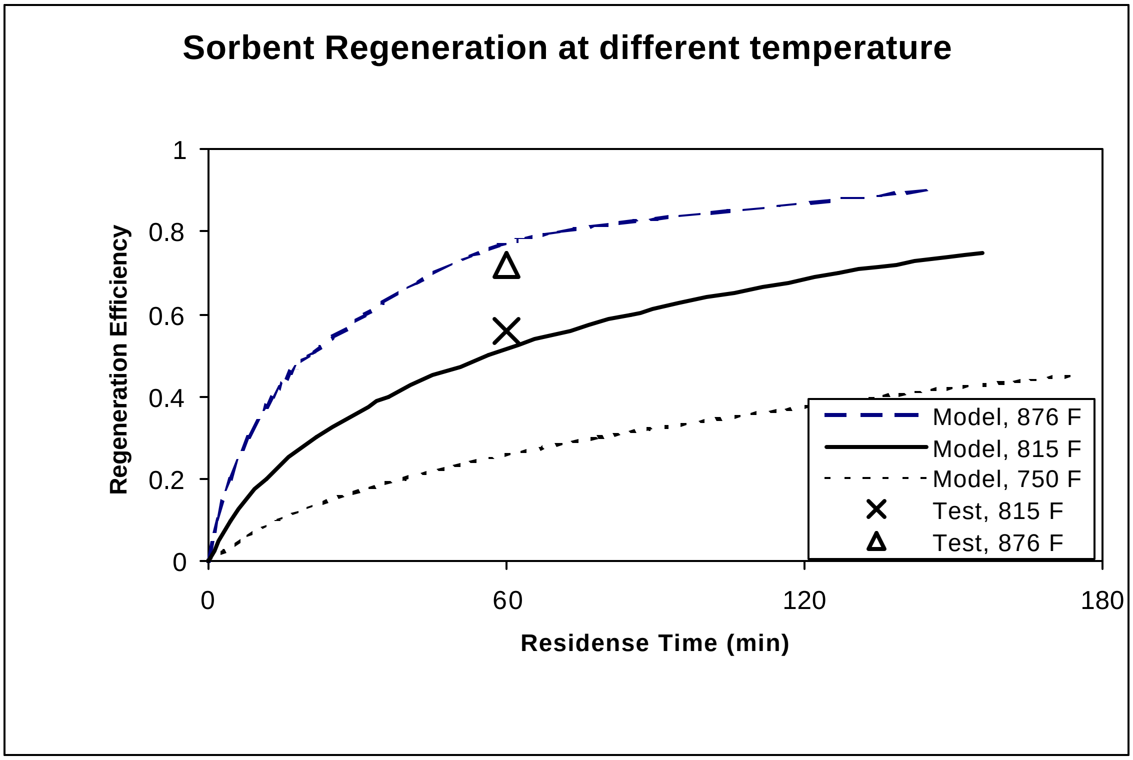


Figure 3. Temperature Dependence in Sorbent Regeneration [9]

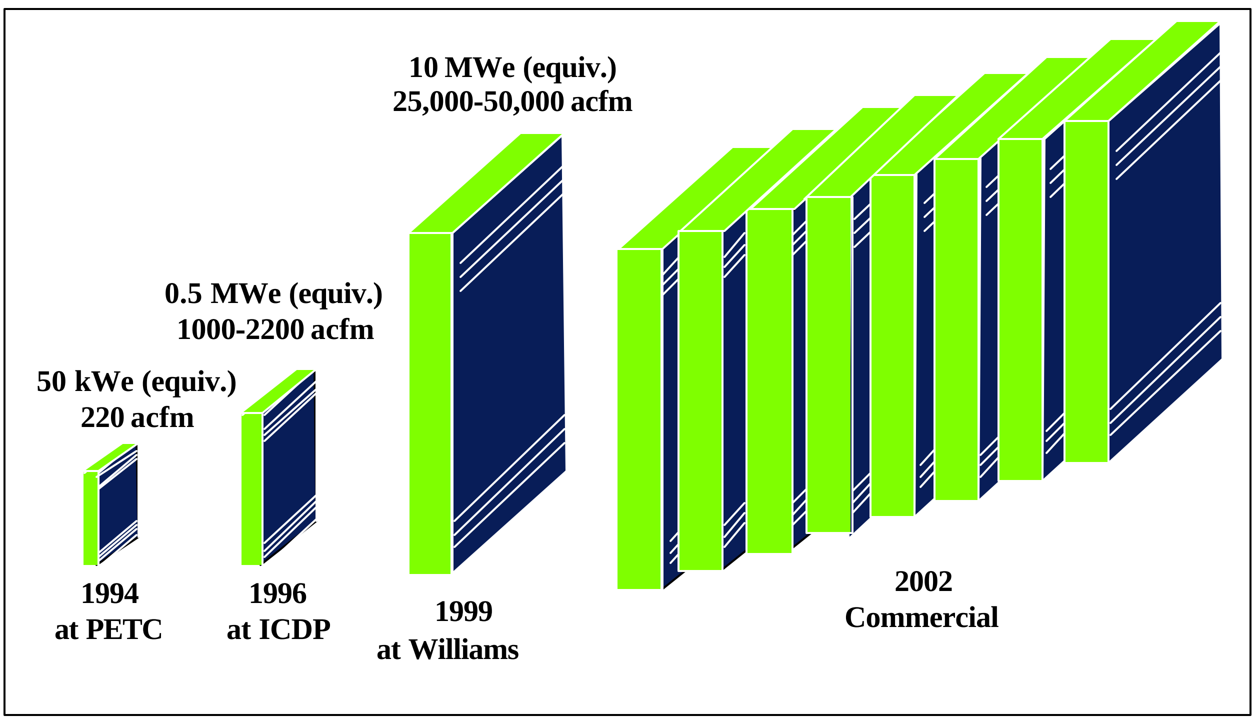


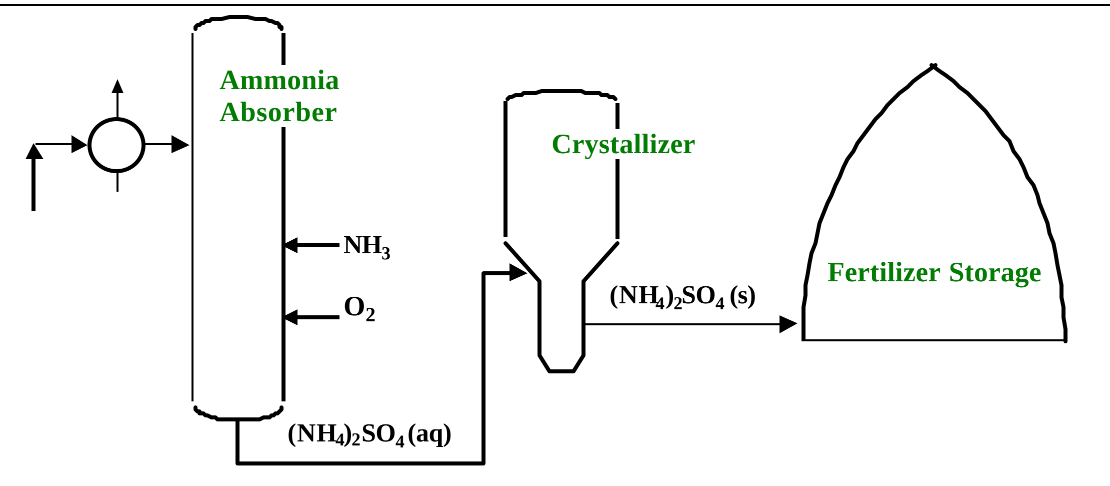
Figure 4. Scale Up Path and Schedule

The other test items include using a new method to produce sorbent. Originally vendors were required to deliver sorbent of gamma alumina spheres impregnated with CuO, and the costs have been around $2-3 /lb. The new way to prepare sorbent is to ask vendors to supply gamma alumina spheres impregnated with $CuSO_4$, and to activate the sorbent in the regenerator. Since $CuSO_4$ impregnated sorbent can be easily prepared by soaking alumina spheres in a $CuSO_4$ solution, the cost of producing sorbent can be dramatically reduced.

Pneumatic and mechanical sorbent transportation methods were tested and pneumatic method was selected. In addition, an ammonia sulfate byproduct facility has been incorporated at the Illinois Coal Development Park. The process is described in the figure below. The final product not only can serve local farming community, but also has a higher economic value than other sulfur-based products. The flexibility in making sulfur based byproduct is one of the major advantage of the Copper Oxide Process. Table 2 gives the approximate value of recovered sulfur by producing different byproducts.

Table 2. Sulfur Recovery Value for Various Byproducts

Byproduct	Byproduct Price per ton	Value per ton SO_2
Sulfuric Acid	$50	$77
Elemental Sulfur	$40	$20
Ammonia Sulfate	$90	$186
Corn Processing Steep Acid	Displace on-site production	To be determined



$SO_2 + H_2O = H_2SO_3$
$H_2SO_3 + 2NH_3 = (NH_4)_2SO_3$
$(NH_4)_2SO_3 + 1/2O_2 = (NH_4)_2SO_4$
Granular $(NH_4)_2SO_4$ ~ $90/ton (~ $186/ton SO_2 removed)

Figure 5. Ammonia Sulfate Fertilizer Byproduct Process

Full Scale Demonstration Test

It is anticipated that full scale copper oxide facilities will be made up of a number of 10MW absorber panels and associated gas and material handling equipment. Several of these modules will be grouped together and will be serviced by one or more regenerators. A full scale absorber module will be around 45 ft high by 10 ft wide by 36 inches deep. The large cross section is mainly due to the large volume of flue gas and selection of an appropriate gas velocity to reduce the pressure drop. The regenerator will be a cylindrical vessel with a smaller size. The sorbent residence time will be approximately 8 hours in the absorber and 3 hours in the regenerator. A sorbent heater may also be required in between the absorber and regenerator to ensure adequate sorbent regeneration, because efficient regeneration requires a temperature around 850 ^{o}F, while the flue gas entering the absorber will be at typical boiler outlet temperatures of 650 to 750 ^{o}F.

A full scale system demonstration will be conducted at Williams Energy Services ethanol plant at Pekin, Illinois. Currently the Williams Plant burns elemental sulfur to produce SO_2. The SO_2 gas is scrubbed with water to form steep acid (H_2SO_3 solution) for preprocessing corn. By using the Copper Oxide Process, the plant can not only reduce the sulfur and NO_x emission from the boiler, but also save costs for purchasing commercial sulfur and burning it to SO_2. This case is a good example showing the advantage of dry regenerable flue gas cleanup process such as Copper Oxide Technology.

An engineering design is scheduled to be completed by the end of this year, and the construction is planned to begin in 2000.

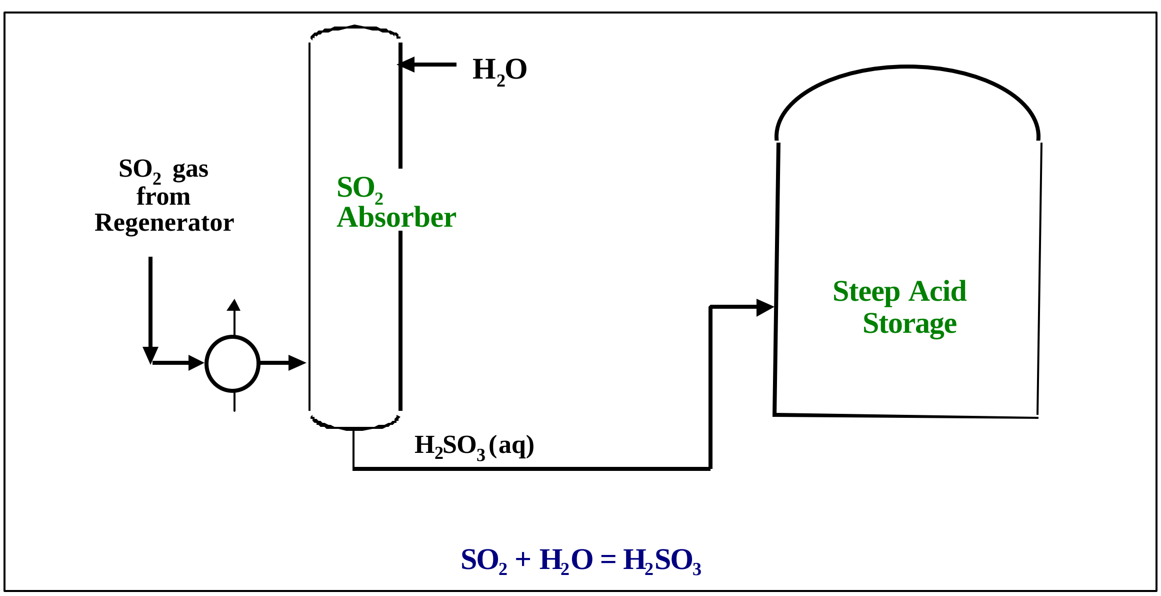


Figure 6. Corn Milling Steep Acid Byproduct Process

Technical Challenges

Although the Copper Oxide Process has been tested for extended periods at small and moderate scales, to develop this technology into a cost effective, utility scale flue gas treatment technology several additional technological challenges must be addressed.

Sorbent and gas flow in a full scale absorber module is one of them. To have a large volume of small sorbent spheres move uniformly in a 45 ft high and 10 ft wide panel is a challenging task. Uneven sorbent flow will either compromise the sulfur removal efficiency or require an unnecessarily high sorbent flow rate. In addition, because the large amount of sorbent is used, it is important to keep the sorbent attrition small. Careful design and testing of pneumatic sorbent transport systems include using appropriate velocity of carrying gas and reducing sorbent damage in valves should be considered.

A related issue is fly ash. An important task is to assure that fly ash will not clog the flue gas flow in the absorber. The ash effect has been tested in smaller units. Because of the variety of ash composition, it will be important the process be tested for an extended period in a full scale unit.

Lastly, because of the competitive nature of electricity generation in the US, and the risk averse mentality in the power industry, a good demonstration of steady performance and low cost are essential for the industry to adopt this new technology. Therefore, after a smooth operation is achieved in the demonstration plant, detailed process economics will have to be evaluated to determine the life cycle operating costs for the technology. Additional cost cutting techniques and methods can also be evaluated and implemented. There are some promising areas that could reduce the capital and operating costs of the technology. A key area is further sorbent development. Sorbents capable of supporting higher copper loading, greater strength, lower pressure drop and faster reaction kinetics need to be investigated. Sorbent formulations capable of supporting catalytic oxidation of methane in the regenerator to permit in-situ sorbent heating or to reduce the required regeneration temperature will also lead to improved process economics.

In order to make steady progress in the scale-up test, the first priority will be to achieve high SO_2 and NO_x removal in a steady operation. After that the focus will be on testing cost cutting measures.

Summary

To develop a regenerable flue gas cleanup that removes multiple pollutants has been a long-term objective of FETC. As a LEBS development team, we are aware that legislative action, public opinion, and electrical generation economics require significant advances in the emission control equipment utilized on coal fired power plants. In the near term starting 2000, the Phase II Clean Air Amendment Act calls for further sulfur reduction in all units, and the NO_x emission will be rigorously controlled by both CAAA and ozone transport rule. For a longer horizon, the US DOE Vision 21 plan calls for steadily further reduction of SO_2 and NO_x emissions to ¼ New Source Performance Standard (NSPS) in 2005 and 1/10 NSPS in 2010[13]. Considering other factors such as the cost and impact of landfill on environment, and the flexibility to use recovered sulfur as valuable byproducts, Copper Oxide Process presents many attractive advantages.

The process has been tested up to 5 MMBTU/hr level and in fixed bed, fluidized bed, and moving bed configurations. These previous tests indicated that the fundamental chemistry of the process is robust. The engineering effort undertaken in this LEBS project is to scale up the process to a scale that will enable commercial acceptance of the technology. Commercial acceptance will require reliable, cost effective operation to be demonstrated to future customers. In addition, the developers of the technology will have to be sufficiently confident in the technology to provide performance and cost guarantees for the commercial systems. Although technical challenges remain, these issues are manageable. Our strategy in the LEBS Phase IV effort will be to keep the high SO_2 and NO_x removal as a first priority, and then to address the cost measures. We believe that by setting up such a strategy, we will be able to achieve a steady progress, and the Copper Oxide Process can serve as a flue-gas cleanup technology for the next generation of coal fired power plants.

Acknowledgement

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